

A review of receptor modelling of industrially emitted particulate matter

Taiwo, Adewale M.; Harrison, Roy M.; Shi, Zongbo

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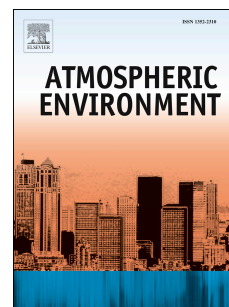
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A Review of Receptor Modelling of Industrially Emitted Particulate Matter

Adewale M. Taiwo, Roy M. Harrison^{*1}, Zongbo Shi

**Division of Environmental Health and Risk Management
School of Geography, Earth & Environmental Sciences
University of Birmingham
Edgbaston, Birmingham, B15 2TT
United Kingdom**

^{*} To whom correspondence should be addressed

Tele: +44 121 414 3494; Fax: +44 121 414 3708; Email: r.m.harrison@bham.ac.uk

¹ Also at: Department of Environmental Sciences / Center of Excellence in Environmental Studies, King Abdulaziz University, Jeddah, 21589, Saudi Arabia

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Highlights:

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- Industrial processes have been identified as an important source of airborne PM.
- PM from different sites within the same industry may vary appreciably in composition.
- PM from different processes within the same industrial site can differ substantially.
- Local source profile measurements are needed for industrial PM source apportionment.

33 This review summarises the different receptor models that have been adopted at industrial and
 34 urban sites to apportion the sources of particulate matter (PM) from industries. Industrial processes
 35 and those associated with industry (such as transportation) are an important source of airborne PM
 36 which includes trace elements, organic and elemental carbon, and PAHs. Industry also emits
 37 gaseous pollutants which form secondary aerosol in the atmosphere. Most published studies have
 38 employed chemical mass balance (CMB), positive matrix factorization (PMF) and/or principal
 39 component analysis (PCA) models as source apportionment tools. These receptor models were
 40 mostly applied to fine particulate matter (PM_{2.5}) and PM₁₀ compositional data, particularly the
 41 inorganic constituents. Some studies have combined two or more of these receptor models, which
 42 provides useful information on the uncertainties associated with different models. Industry has been
 43 reported to contribute from 0 to 70% of PM mass at industrial sites. It appears that some studies are
 44 unsuccessful in apportioning PM from industry, e.g., unable to distinguish industrial emissions from
 45 other sources. A critical evaluation of the literature data also showed that the choice of appropriate
 46 tracers for industry, both generically and for specific industries, varies between different PM source
 47 apportionment studies. This is not surprising considering the significant difference in source
 48 profiles of PM from different types of industry, which may compromise source apportionment of
 49 industrial emissions using CMB with non-local source profiles. It may also affect the attribution of
 50 industrial emissions in multivariate statistical models (e.g. PMF and PCA). It is concluded that a
 51 general classification of the source “industry” is rarely appropriate for PM source apportionment.
 52 Indeed, such studies may even need to consider the different processes within a particular industry,
 53 such as a steelworks, which emit PM with significantly different chemical signatures. It is suggested
 54 that future source apportionment studies should make every effort to measure source profiles of PM
 55 from different industrial processes, and where possible, use multiple models in order to more
 56 accurately apportion the source emissions from industry.

57 **Keywords:** Source apportionment; industrial emissions; receptor modelling; metals; particulate
 58 matter; steel industry

60 Airborne particulate matter (PM) is a complex pollutant emitted directly from anthropogenic and
61 natural activities (Poschl, 2005) or formed indirectly as secondary aerosol (Harrison and Yin, 2000).
62 Particulate pollutants are composed of a complex mixture of substances with diverse physical,
63 chemical and biological composition. A number of health problems have been associated with
64 exposure to PM. For example, epidemiological studies have found strong correlations between
65 concentrations of PM and hospital admissions and mortality due to respiratory and cardiovascular
66 diseases (Pope and Dockery, 2006).

67
68 Rapid economic and industrial developments have led to increased energy consumption, emissions
69 of air pollutants and poor air quality in major cities of the world, especially in developing countries
70 (Chan and Yao, 2008). Hence, there is a compelling need for quantification, identification and
71 apportionment of these pollutants in order to facilitate their reduction through proper management
72 plans. Emission inventories and chemistry-transport models are important tools in the evaluation of
73 particulate matter pollution. However, these models have some limitations, e.g., due to the fact that
74 important sources of PM are fugitive and hence often poorly quantified (Almeida et al., 2005).
75 Additionally, gas-to-particle transformation models are not always able to describe adequately the
76 contribution from secondary aerosol. Therefore, receptor modelling remains an important tool.
77 Receptor modelling uses physical and chemical characteristics of air pollutants to identify and
78 apportion their contributing sources. The two generic method types of receptor models are the
79 Chemical Mass Balance (CMB) model (Watson et al., 2002) and factor analytical methods (Hopke,
80 2003). The latter includes Principal Component Analysis-Absolute Principal Component Scores
81 (PCA-APCS) (Thurston and Spengler, 1985; Garcia et al., 2006), Positive Matrix Factorization
82 (PMF) (Paatero and Tapper, 1994) and UNMIX (Henry, 1997, 2002).

83
84 Application of receptor models for source apportionment of PM has been well established in
85 published pollution studies (Viana et al., 2008a; Yatkin and Bayram, 2008; Mansha et al., 2012;

86 Belis et al., 2013, 2014; Pant and Harrison, 2012; Pant and Harrison, 2013). Industry has frequently
87 been reported to be one of the important sources of airborne PM, alongside other sources such as
88 traffic, crustal material, secondary aerosol, sea spray, incineration, fuel oil burning, biomass
89 burning, and coal combustion (Harrison et al., 2003; Marcazzan et al., 2003; Qin and Oduyemi,
90 2003; Chio et al., 2004; Karar and Gupta, 2007; Pant and Harrison, 2012), particularly in industrial
91 cities (Oravisjarvi et al., 2003; Querol et al., 2007; Tsai et al., 2007; Alleman et al., 2010). In EU
92 member states, industrial processes are the second and third largest source of primary PM_{2.5} and
93 PM₁₀ respectively ([http://www.eea.europa.eu/data-and-maps/figures/sector-contributions-of-](http://www.eea.europa.eu/data-and-maps/figures/sector-contributions-of-emissions-of-2)
94 [emissions-of-2](http://www.eea.europa.eu/data-and-maps/figures/sector-contributions-of-emissions-of-2)). The objective of this review is to examine critically the application of various
95 receptor models used for source apportionment of particulate matter from industry. It will first give
96 an overview of the emitted PM pollutants from industries and particularly the iron and steel
97 industries. It will then review the receptor modelling of PM from industries. Finally it will compare
98 the PMF profiles used in the literature with the USEPA SPECIATE database for different processes
99 for iron and steel-making in order to evaluate the results from current receptor modelling on
100 industrial pollutants from this source.

101

102 **2. INDUSTRIAL EMISSIONS OF PARTICULATE MATTER POLLUTANTS**

103 In this section, the major types of PM-related pollutants will be briefly introduced. It will also
104 include a specific exemplar industry, iron and steel manufacturing, which has been the subject of
105 several studies.

106

107 **2.1 Major Particulate Phase Pollutants from Industries**

108 There are many types of industry. Major primary industries that contribute to PM emissions include
109 but are not limited to: manufacturing (including automotive, steel and metal-making industries),
110 aerospace, agriculture, chemical, construction, energy. Particulate phase pollutants from industries
111 include metals (Querol et al., 2007; Cetin et al., 2007), OC/EC (organic carbon, elemental carbon),
112 PAHs (Rehwagen et al., 2005; Jang et al., 2013), and water soluble ions (Querol et al., 2002).

114 Trace metals are one of the most characteristic chemical species associated with PM emission from
115 many industries and are the major tracers used in many receptor modelling studies. Trace elements
116 in industrial PM emission are related to the handling and processing of raw materials, handling of
117 intermediate products and production of end products.

118
119 EEA (2012) reported that industrial processes make a significant contribution to the total EU-27
120 emissions of heavy metals (36% Pb, 25% Cd, and 41% Hg), despite significant reductions since
121 1990. In Canada, industrial processes account for 72%, 79% and 32% of total emissions of Pb, Cd
122 and Hg (<http://www.ec.gc.ca/inrp-npri/default.asp?lang=En&n=0EC58C98-1>). Metal production is
123 one of the major industrial processes contributing the emissions of total trace elements such as Cd,
124 Cr, Cu, Hg, Ni, Se, V, and Zn in the UK in 2009 (Table 1). A study by Querol et al. (2007) also
125 showed a number of trace elements in airborne PM are associated with industrial emissions in
126 Spain.

127
128 Analysis of airborne PM close to steel plants has shown that Fe, Mn, Zn, Pb, Cd and K are
129 associated with emissions from the steel and iron plants. Microscopic analysis of individual
130 particles has confirmed the presence of individual Fe-rich particles close to steel plants. For
131 example, Moreno et al. (2004) identified iron spherules in both fine and coarse PM fractions at a
132 steelworks in Port Talbot, South Wales, UK; Ebert et al. (2012) observed a significant fraction of
133 individual iron oxides and iron mixtures in airborne PM near a steel industry in Duisburg, Rhine-
134 Ruhr area, Germany. Elevated concentrations of some elements at the steel industry sites derive
135 from the raw materials being used for steel production. For example, raw materials including iron
136 ores (FeO , Fe_2O_3 , Fe_3O_4), limestone (CaCO_3) and dolomite ($\text{CaMg}(\text{CO}_3)_2$) are used in a blast
137 furnace (BF) while lime (CaO) and fluorspar (CaF_2) are used in a Basic Oxygen Steel plant
138 (Machemer, 2004). Integrated steel plants are also known for high emissions of mercury (Pacyna
139 and Pacyna, 2002; Themelis and Gregory, 2002; Borderieux et al., 2004). Asia and Europe are the

regions where steel industries contribute most to the global mercury budget (Pirrone et al., 2001; Pacyna et al., 2006). Mukherjee et al. (2008) reported that annual mercury emissions from iron and steel industries in India increased by 25% between 2000 and 2004.

2.1.2 Organic/elemental carbon (OC/EC)

Carbonaceous particles comprising OC and EC are another pollutant generated from industrial emissions. Some of the OC/EC are directly emitted from particular industrial processes and some are associated with relevant activities of industries (see Section 2.3). Globally, industries contribute about 10% and 15% of OC and BC (black carbon) emissions respectively (Bond et al., 2004). In early 21st century China, industrial BC emissions arise primarily from uncontrolled coal-fired stokers and from the production and use of coke in the iron and steel industry; total coal-derived emissions are 83 Gg (85% of the industrial sector total of 97 Gg) (Streets et al., 2001).

Tsai et al. (2007) measured elevated concentrations of OC and EC at the cold forming unit of an integrated steelworks in southern Taiwan. Highly time-resolved ambient measurements made at a fence-line site adjacent to a large coke production plant in the USA also revealed high concentrations of OC (40% of total measured $PM_{2.5}$) and EC (25% of measured $PM_{2.5}$) (Weitkamp et al., 2005).

Polycyclic Aromatic Hydrocarbons (PAHs) are a group of organic compounds that are mainly produced by incomplete combustion and pyrolysis of organic material (Manahan, 2009). Industrial processes are a minor source (5%) of PAHs in the UK in 2008 (AEA, 2010) and contributes 9% of PAHs in the EU in 2011 (EEA, 2012). The most important industrial sources of PAHs include primary aluminium and coke production (e.g. as part of iron and steel production), waste incineration, cement manufacture, petrochemical industries, creosote and wood preservation, bitumen and asphalt industries, rubber tyre manufacturing, and commercial heat/power production (European Commission, 2001) and paper mills (Fauser et al., 2011). The PAH emission factors are

affected by incoming fuel, the manufacturing process, and the air pollution control devices (Ravindra et al., 2008). The significantly higher PAH levels at an industrial site than at control site and the La Plata city centre in Argentina also suggest that the refinery and petrochemical plants are important sources of PAHs (Rehwagen et al., 2005).

2.1.3 Water soluble ions

Water soluble ions have been observed at significant concentrations in industrial sites (Kumar et al., 2001; Oravisjarvi et al., 2003; Samara et al., 2003; Karar and Gupta, 2007; Gildermeister et al., 2007; Viana et al., 2008b; Amato et al., 2009; Zeng et al., 2010; Pancras et al., 2013). K and Cl have been suggested to be associated with sinter plant emissions (Dall'Osto et al., 2008; Hleis et al., 2013).

2.2 PM Pollutants from Different Processes within a Particular Industry: Steel manufacturing as an example

A modern integrated plant is usually a complex operation, with more than one industrial process unit. For example, steel manufacture involves coke production, sintering, blast furnaces and basic oxygen furnace steelmaking (BOF). Sintering involves mixing iron ores, filter dusts and mill scale all fused together as appropriate feedstock for the BF (Brigden et al., 2000), while in coking processes, pulverized bituminous coal is used as fuel in order to reduce iron oxides and remove volatile impurities (<http://ecm.ncms.org/ERI/new/IRRironsteel.htm>). The BF is a high temperature driven process where metallic iron reduction from the oxide form takes place by burning with coke produced in the coking process. The steelmaking section involves addition of various alloying elements to give the finished materials the combination of properties desired. This takes place in three ways, i.e. the BOF for processing pig iron and the electric arc furnace (EAF)-for recycled materials and the open hearth furnace (OHF) where excess carbon and other impurities are burnt out of the pig iron to produce steel. Presently, about 70% of the world steel is produced from BOF while 29% comes from EAF (<http://www.worldcoal.org/coal/uses-of-coal/coal-steel/>).

Each unit may emit PM with specific characteristics. Figure 1 shows the source profiles of PM from two industrial processes in a steel plant and charcoal manufacturing from the USEPA SPECIATE database. There are significant differences in the emission profiles. For example, BOF with an electrostatic precipitator produces high concentrations of sulphate and Si, whereas BOF without control emits high concentrations of Fe and Mn but lower concentrations of sulphate and Si; a sinter plant generates high emissions of Fe, Pb, K and Cl, whereas charcoal manufacturing (not necessarily for a steel plant) has a high concentration of Al, Ca, and Se (Figure 1). A recent study by Tsai et al. (2007) also suggested that K and Pb, which contribute a significant percentage (15 and 2 %) to the total observed particle mass, are associated with the sintering process. Similarly, Oravisjarvi et al. (2003) found that the sinter plant contributes 96% and 95% of the total measured concentrations of Pb and Cd in PM at Rahee, Finland. The study of Machemer (2004) showed elevated concentration of Fe, Al, Si, S and Zn at sections close to both BOF and blast furnaces (BF). At the coke making process, major elements observed by Tsai et al. (2007) were S, Fe and Na. In the cold forming aspect of the iron and steel industry, major elements observed in the particles were S, Fe, Na, K and Ni. The hot forming process showed a high abundance of S, Fe, Na and Ca (Tsai et al., 2007). These reports highlight the importance of using local profiles for CMB type models, and provide useful references for identifying tracers for factor analysis-based models. More detailed discussion on these aspects will be given later.

2.3 PM Pollutants from Industrially Related Activities

When considering PM emission from a particular industry, one should also consider the other processes that are associated with that industry, for example, transportation of raw materials and end products and energy consumption. This leads to primary emission of combustion aerosols, including vehicular emissions as well as re-suspended dust. Based on emission reports by EU countries under the CLRTAP and NEC Directive, EEA (2012) estimated that 36%, 25%, and 41% of the Pb, Cd and Hg in the EU is emitted from energy use in industry. It is however challenging to apportion the emissions from energy use in industry relative to non-industrial sources. It is also

difficult to distinguish the re-suspended dust from the industry itself and those from other processes such as wind-blown dust and the dust generated from working agricultural land. Dust resuspension from raw material transportation is especially relevant in the case of the ceramic industry.

Apart from the primary particulate pollutants discussed above, industries are also known for emission of gaseous pollutants such as carbon monoxide (CO), sulphur dioxide (SO₂), nitrogen oxides (NO_x) and hydrogen gas (H₂), and volatile organic carbon (Ogulei et al., 2006; Ogulei et al., 2007; Tsai et al., 2008; Johansson and Söderström, 2011; Pancras et al., 2013). Some of these gaseous pollutants can be transformed into secondary compounds which are commonly detected in urban aerosols. It is very challenging for receptor modelling to estimate how much of the secondary aerosols are from the primary pollutants emitted from different industries.

3. RECEPTOR MODELLING OF PM FROM INDUSTRIAL EMISSIONS

3.1. Industrial Sites

Table 2 summarizes different methods of receptor modelling applied to ambient PM measurements at industrial sites all over the world. The types of industries where receptor monitoring sites were located include steelworks, metallurgical plants, oil refineries, petrochemical works and small factories. Most of the studies collected PM₁₀ or PM_{2.5} samples, with a few also collecting data on total suspended particles (TSP) or PM_{2.5-10}. Trace metal concentrations were often used as the source data for receptor modelling (Table 2). OC/EC and ionic components were also included in combination with the metals in some studies. A few of the studies included PAHs as source data. Several types of models have been used including PCA, PCFA (Principal Component/Factor Analysis), CMB, Nested CMB, PMF or Multilinear Engine-2 (ME-2).

Elevated PM mass concentrations have been observed at some industrial sites compared to residential stations. For example, Xue et al. (2010) reported that the annual PM₁₀ concentrations ranged from 131 to 179 $\mu\text{g m}^{-3}$ at industrial sites compared to 86 $\mu\text{g m}^{-3}$ at a rural background site

in Panzhihua, China. Kim and Jo (2006) showed that the average PM_{10} mass levels was 81 and 71 $\mu g m^{-3}$ during winter and summer at an industrial site in Pohang, Korea compared to 52 and 42 $\mu g m^{-3}$ observed at a residential site. Yatkin and Bayram (2008) found that PM_{10} mass levels were 80 $\mu g m^{-3}$ at an industrial site, which is about twice that of a suburban site in Izmir, Turkey. Querol et al. (2006) reported that the PM_{10} mass level at an industrial site (Changqian, China) was 197 $\mu g m^{-3}$, which was 41 $\mu g m^{-3}$ higher than that at an urban site (Hankou, China). An extremely high PM_{10} concentration, 305 $\mu g m^{-3}$, was also reported at an industrial site in China (Zeng et al., 2010). It is apparent that many of these studies conducted at both industrial and residential/background/urban sites report mass levels of PM_{10} greater than the European Union 24-hour mean Limit Value of 50 $\mu g m^{-3}$.

The sources (factors) identified in different studies include industry, fuel/oil/coal combustion, traffic (including exhaust and non-exhaust emissions), crustal (soil/dust/minerals), secondary, marine and waste incineration. Literature reports have identified many different types of sources, some of which are similar but with different terminology (e.g., Kim and Jo, 2006; Querol et al., 2006; Ogulei et al., 2006; Viana et al., 2008; Lim et al., 2010). To simplify the comparisons, we also combined some of the sources together to report in Tables 2 and 3. The details of such combinations are shown in the footnotes of Tables 2 and 3. If one or more sources are not classified into one of the categories in Tables 2 and 3, they are listed as “others”. “Others” also include mixed sources such as metallurgy/fossil fuel combustion and waste incineration/marine aerosol by Kim and Jo (2006), steel and fuels by Yatkin and Bayram (2008), incineration and Pb-related industry by Lim et al. (2010), and regional and marine by Viana et al. (2008b); vehicle and industrial oil burning by Lodhi et al. (2009).

In some cases, up to 48% of the source contributions were not identified, suggesting that the model resolution was not good enough. This could be due to inadequate data (e.g. insufficient samples, poor quality analytical data or inappropriate sets of analytes) causing a failure of the modelling.

275 A few studies have adopted two or more receptor modelling approaches for source apportionment.
276 Some of them produced similar results from different models, such as Viana et al. (2008) and
277 Callen et al. (2009). However, source contributions estimated in some studies (e.g., Yatkin and
278 Bayram, 2008; Srivastava and Jain, 2008) are significantly different using different models. In the
279 latter cases, it is possible that one of the models or both failed to produce satisfactory resolution
280 and/or that the datasets are insufficient to resolve the sources. For application of the multivariate
281 statistical models (PCA, PMF) it is strongly advisable for the ratio of the number of independent
282 samples to the number of species entered in the model to exceed three (Thurston and Spengler,
283 1985), but this guideline is not observed in all cases, leading potentially to model instability.

284 The reported contribution of industry to the PM mass is highly variable. Most studies have
285 apportioned less than 10% of PM to industrial sources (Table 2). A very low industrial contribution
286 of 1% has been reported (Ogulei et al., 2006; Gupta et al., 2007). In one case, no contribution from
287 industry was identified at an industrial site (Samara et al., 2003). In Hammond et al. (2008), iron–
288 steel manufacturing/waste incineration together contributed 0.1 and 4% to PM_{2.5} at East and
289 Southwest Detroit, respectively. In Callen et al. (2009), industry and traffic sources were identified
290 as a single factor. In these studies, the specific contribution from industry cannot be ascertained. It
291 is clear that in some studies, the contribution of PM from “industry” is beyond the resolution
292 capabilities of the RMs used (contributions of 1-2% or less). In Lodhi et al. (2009), the contribution
293 to PM_{2.5} from the steel industry was resolved to be 8% but the full contribution from industries must
294 be more than 8% because another mixed source include industrial emissions. Several studies have
295 apportioned more than 10% of PM to industry (Alleman et al., 2010; Viana et al., 2008b; Chung et
296 al., 2006; Yatkin and Bayram, 2008; Karar and Gupta, 2007; Oravisjarvi et al., 2003). The highest
297 contribution from industry to ambient PM reviewed in this study was estimated to be 70% by Cetin
298 et al. (2007).

299

300 There are many reasons for the large difference in the apportioned contributions of PM from
301 industry including: (i) distance of the industrial units to the sampling sites; (ii) meteorological

conditions (e.g. whether the site is downwind or upwind); (iii) particulate emission control measures in place in most industrial plants. Another potentially crucial reason is the choice of industrial tracers in the receptor models in these studies. Multiple emission sources of certain marker elements could create conflicts during apportionment exercises. This will be discussed in more detail in the next section.

The contribution of combustion sources, encompassing stationary burning of oil, fuel, wood or coal, to the total PM ranges from 0.4 to 58% (Table 2). Combustion is reported to be a major source (>20%) of PM in some of the studies (e.g. Xue et al., 2010; Karar and Gupta, 2007; Querol et al., 2006; Mazzei et al., 2008; Chung et al., 2006; Samara et al., 2003; Castanho and Artaxo, 2001). Nested CMB applied for source apportionment of PM₁₀ by Xue et al. (2010) at industrial sites in China indicated coal combustion ash to represent the largest source of PM₁₀ (26%). However, it needs to be emphasized that the influence of combustion sources on ambient PM may or may not be directly related to industry. No contribution from combustion was identified in a few studies (Oravajarvi et al., 2003; Cetin et al., 2007; Viana et al., 2008b; Yatkin and Bayram, 2008; Alleman et al., 2010). This could be due to an insignificant contribution from the combustion sources but model bias or incapability of the models to identify the source could not be ruled out. Jang et al (2013) found a similarity in the PAH congener profiles of coal combustion and steel industry emissions, which was resolved only by inclusion of a large number of congeners.

The traffic source is often a major source of PM even at the industrial sites. It is typical of industrial areas to have high traffic flows due to transportation of raw and processed goods as well as personnel mobility. Heavy-duty vehicles, known large emitters of particles (Charron and Harrison, 2005), are often used for transportation of raw materials and processed goods in industry. This is in addition to the contribution from vehicular emissions not associated with the industry and long-range transported sources at those sites.

Particles with a crustal signature comprised of soil and road dusts as well as cement dust are another important source of PM at industrial sites. Most studies listed in Table 2 have attributed an appreciable proportion of PM, especially in $PM_{2.5-10}$, PM_{10} and TSP fractions, to crustal matter. Resuspended dusts from roads and all forms of construction works, and windblown soil at the industrial sites are probable contributors. Vehicular movements at the industrial sites could increase dust particles through resuspension processes (Charron and Harrison, 2005). However, it is difficult to apportion the crustal matter from industry due to the overlapping signatures of the possible contributing sources.

3.2 Urban/Residential Sites

As stated above, PM mass concentrations in a number of selected studies in residential areas were generally lower compared to the values reported at the industrial sites. Table 3 compiles selected source apportionment studies carried out in urban/residential areas. Despite the fact that studies presented in Table 3 were conducted around the perimeter of residential/urban areas sites, an industrial factor was still prominent in some of the receptor modelling studies. The percentage contributions assigned to industry range between 2 and 37%. Elevated percentages assigned to the industrial source in the residential areas might be related to prevailing meteorological conditions (wind direction) during sampling, and the source to receptor site distances maximising the impact through local dispersion processes (e.g. from an elevated point source) (Almeida et al., 2005; Yatkin and Bayram, 2008).

Table 3 also shows that some studies were unable to differentiate between industry and traffic by the receptor models used and therefore reported them as a single source (Almeida et al., 2005; Callen et al., 2009). Coal combustion is another significant source of PM pollution in the residential areas, especially in studies from India and China (Chowdhury et al., 2007; Xue et al., 2010). This may arise partly from industrial processes.

357 **INDUSTRIAL EMISSIONS**

358 The choice of marker elements for industrial factors/sources in source apportionment is a crucial
 359 aspect of receptor modelling. In source apportionment studies, different authors have chosen
 360 different marker elements for “industry” (as a general term), for example: As, Zn, Pb, Cs, Tl, Zr,
 361 Hf, Ce and Cu (Almeida et al., 2005; Viana et al., 2008b); Pb, Co, Ce, Cr (Hien et al., 2001); Zn,
 362 Pb, Si, Ni, Mn, Fe, S (Castanho and Artaxo, 2001); Cd, Pb, Cr, Ni (Heal et al., 2005); Zn and Pb
 363 (Zn and Pb smelters, Connell et al., 2006; Kim et al., 2007; Mazzei et al., 2008). It needs to be
 364 emphasized that “industry” here refers to a general term rather than a specific industry. A more
 365 comprehensive list of marker elements attributed to industrial sources is shown in Tables 2 and 3.

366
 367 A range of marker elements have been used for the steel industry. For example, in the study of Xue
 368 et al. (2010) at a mixed industrial location with iron and steel industries, Ti, Cr and Mn were used
 369 as markers for metallurgical industry. Tsai et al. (2007) and Oravisjarvis et al. (2003) used K, Pb,
 370 Fe, Ca, S/SO₂ and Na as tracers for steel production. In the work of Cetin et al. (2007), Zn, Fe, Pb,
 371 Mn and Cd were used as steel industry fingerprints. The study by Hammond et al. (2008) adopted
 372 Zn, Fe, Mn, K and Pb as steel emission tracers, some of which are also used by other authors.

373
 374 There are several major issues arising from the above discussion on the choice of tracer elements
 375 for industries. The first issue is how “industry” is defined. “Industry” is in general referred to as one
 376 category of emission sources which encompasses a wide range of plants. It is clear that “industry”
 377 in one study is not necessarily the same as that in another study. In some cases, there is only one
 378 dominant industry that may affect the PM in a particular area. This should to some extent facilitate
 379 the choice of tracer elements for receptor modelling. However, frequently, there is more than one
 380 industry in a particular area. Since each industry may emit PM with sharp differences in source
 381 profiles, using a single set of tracer elements to distinguish the source contribution from “industry”
 382 as a whole can be problematic.

The second issue is related to different emission source profiles from industrial processes in a particular industrial unit. Marker elements for industries depend on the nature of different processes and activities taking place within the industry (Querol et al., 2007). As shown in Figure 1, different processes at a steel and iron plant can have significant differences in their source profiles. Therefore, using a single set of elements as tracers for a particular industry as a whole can sometimes be problematic as well.

The third issue is the presence of abatement plant for a particular industrial process in a particular industrial unit. For example, the source profiles of BOF with an electric precipitator are significantly different to those without control (Figure 1). This may to some extent support the choice of different tracer elements for a particular industrial process in different studies but this choice needs to be justified by actual source profile measurements.

The fourth issue of concern is that the multiple sources of some elements that have been used as marker elements may lead to wrong attribution of a source. For example, K is emitted from burning wood or other biomass, vehicular sources, sinter plants and incinerators (Hays et al., 2005; Lim et al., 2010; Hleis et al. 2013). Fe is a component of crustal matter and Fe, Cu, Zn, and Ba are associated with non-exhaust emissions from road traffic (Thorpe and Harrison, 2008; Pant and Harrison, 2013). OC/EC are emitted from many sources including road traffic. Calculation of enrichment factors (EF) can be useful to differentiate natural and anthropogenic emissions (Kothai et al., 2011). The ratios of some specific elements may also be employed to differentiate steelworks emissions from either traffic or other anthropogenic sources. Connell et al. (2006) used the Mn/Zn ratio to identify steelworks emissions in PM_{2.5} sampled at Steubenville, OH, USA. Cl and S are sometimes used as tracers for industries (Prati et al., 2000; Chung et al., 2006) but there are obviously many other potential sources for these elements. It is therefore difficult to resolve the emission sources of elements such as K, Cl, S and Fe in receptor modelling except by inclusion of

other tracers. A typical example is the use of levoglucosan along with K as tracers for biomass burning (Zhang et al., 2010; Harrison et al., 2012).

The summary in Tables 2 and 3 shows a wide range of tracer elements for “industries”. This to some extent is justifiable because different industries have different chemical signatures (tracers). Many previous receptor modelling studies appear to over-simplify the source apportionment of industrially emitted PM. There is a tendency in some studies using PMF or PCA methods to associate factors containing trace metal signatures with “industry” without supporting information on industrial emission profiles. We recommend that the choices of tracers for industries should be supported and justified by comprehensive source profiles from major industries in the area of interest.

In the following, we will quantitatively compare the profiles of PMF factors used for source apportionment of PM from steel production processes with those of the USEPA SPECIATE source profile. Our intention is to further examine the appropriateness of the choice of tracer elements for different processes associated with steel industry activities.

5. PMF FACTORS VERSUS USEPA SPECIATE PROFILES FOR STEEL PRODUCTION PROCESSES

Here, we compare the factor profiles identified by PMF with USEPA SPECIATE source profiles from general steel production processes, sintering and coking processes and a blast furnace (Figures 2a-2d). We have chosen these processes mainly because of the data availability (both the USEPA source profiles and the PMF factors). Only a few PMF studies have identified the steel industry and/or process units related to the steel industry as a source. Some studies have identified a steel industry factor but this was mixed with another source such as waste incinerator. In this latter case, the factor was not included in the comparison in Figure 2. It should be emphasised that

these PMF factors are different from ambient concentrations of PM components and represent the fractional composition of the factor attributed to the specific source.

Figure 2a shows that there are some differences in the PMF profiles of steel production with general iron production source profiles in the USEPA SPECIATE database, but most of the elements and in particular the tracer elements, including Fe, Zn and Mn, fall within the 1:10 and 10:1 lines. Some elements such as Ni, Cr, Cu and sometimes K are outside of the 10:1 lines. This may be linked to their use in specific processes not represented in either the PMF or SPECIATE profiles. The level of information associated with each dataset is insufficient to make a specific judgement.

Figure 2b shows that the Fe content in the PMF profile of the sinter plant factor from Alleman et al. (2010) is similar to that in USEPA SPECIATE database. However, most of the other elements are outside of the 1:10 line. In terms of coking, there seem to be large discrepancies between the Alleman et al. (2010) PMF factor profile with that in the USEPA SPECIATE database (Figure 2c). This may result from differences in the trace element composition of feedstocks for the respective plants that were sampled. The PMF blast furnace factor (Steel 1) profile from Taiwo et al. (2014) agrees very well with the USEPA SPECIATE profiles from the blast furnace process (Figure 2d). Fe, K, Mn, and Zn are close to the 1:1 line, while Ni and Cu were outside of the 1:10 line and Cr was not identified in the PMF factor.

In the USEPA SPECIATE/PMF scatter plot shown in Figure 2(a-d), good agreement was observed for some marker elements adopted in different studies, suggesting that the choice of relevant factors was appropriate. However, some discrepancies were also observed for tracers related to steelworks processes. This may be caused by several factors.

One reason may be differing materials inflow at the steelworks processing units reported in the USEPA SPECIATE database and the published work, which may result in a dissimilar chemical profile of PM emissions from each process from plant to plant. Unfortunately, there are limited source profile data available in the literature to evaluate this possibility. The relationship of the source profile measured directly upon emissions from the sintering process from Tsai et al. (2007) against USEPA SPECIATE data is shown in Figure 3. Most elements are within 1:10 and 10:1 lines except for Cu and Ba, which gives some confidence in the comparison of source profiles from different studies. However, there are clear differences for many of the elements.

A second reason may be related to pollution control systems in place at some steelworks. On the issue of pollution control systems, the USEPA SPECIATE data showed that there are significant differences in source profiles of PM from BOF with and without an electrostatic precipitator (Figure 1). Information on the pollution control system at the steelworks foundry, sintering and EAF are not revealed in the USEPA SPECIATE database. However, no control system was in place at the coking plant represented in the USEPA SPECIATE data.

These above discussions indicate that even with the PMF receptor modelling, it is preferable to obtain the source profiles from on-plant measurements to support the choice of factors and tracer elements.

7. CONCLUSIONS AND RECOMMENDATIONS

Industrial emissions are an important source of particulate substances including metals, carbonaceous species and PAHs in the atmospheric environment. Receptor models such as CMB, PMF and PCA have been used to quantify the contribution of industrial emissions to ambient PM. A few studies have combined two or more models which makes it possible to compare the performance of each model. Receptor modelling at industrial sites has assigned between 0 and 71% of PM to industrial emissions. This assignment does not generally include the likely contribution of

industrially-related activities such as combustion and transportation of materials. A comprehensive evaluation of different receptor modelling studies at industrial sites demonstrated that many different elemental profiles have been attributed to industry, often without any check against known source profiles for local industrial processes. This makes it difficult to evaluate the results from these source apportionment studies, in particular when more complete information including the control technologies at the plants and the source profiles of PM from the industries around the receptor sites is not available.

We suggest that in future receptor modelling of industrially emitted PM:

- (1) Where possible, multiple receptor modelling techniques are used in order to provide a means to evaluate the uncertainties.
- (2) Receptor modelling at paired sites, one close to an industrial site and a background site, are conducted to allow a quantitative evaluation of the impact of a particular industrial plant.
- (3) Local source profiles of PM from different industries which may contribute to the receptor site are measured to support the assignment of source profiles in non-CMB type modelling techniques.
- (4) More source profiles associated with different industrial processes in different industries should be measured to enhance the data available for use in CMB models and to assist source attribution in PCA, PMF and related models.

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REFERENCES

AEA, 2010. UK Emissions of Air Pollutants 1970 to 2008. UK Emissions Inventory Team, Department for Environment, Food and Rural Affairs.

Amato, F., Pandolfi, M., Escrig, A., Querol, X., Alastuey, A., Pey, J., Prez, N., Hopke, P. K. 2009. Quantifying road dust resuspension in urban environment by Multilinear Engine: A comparison with PMF2. *Atmospheric Environment* 43, 2770-2780.

Alastuey, A., Moreno, N., Querol, X., Viana, M., 2007. Contribution of harbour activities to levels of particulate matter in a harbour area: Hada Project-Tarragona Spain. *Atmospheric Environment* 41 (30), 6366-6378.

Alleman, L. Y., Lamaison, L., Perdrix, E., Robache, A., Galloo, J. C., 2010. PM10 metal concentrations and source identification using positive matrix factorization and wind sectoring in a French industrial zone. *Atmospheric Research* 96, 612–625.

Almeida, S. M., Pio, C. A., Freitas, M. C., Reis, M. A., Trancoso, M. A., 2005. Source apportionment of fine and coarse particulate matter in a sub-urban area at the Western European Coast. *Atmospheric Environment* 39, 3127-3138.

Belis, C.A., Karagulian, F., Larsen, B.R., Hopke, P.K., 2014. Critical review and metal-analysis of ambient particulate matter source apportionment using receptor models in Europe. *Atmospheric Environment*. 69, 94-108.

Belis, C.A., Larsen, B.R., Amato, F., Haddad, I.E., Favez, O., Harrison, R.M., Hopke, P., Nava, S., Paatero, P., Prevot, A., Quass, U., Vecchi, R., Viana, M., 2014. European guide on air pollution source apportionment with receptor models. JRC reference report, Report EUR 26080 EN.

Bhanuprasad, S. G., Venkataraman, C., Bhushan, M., 2008. Positive matrix factorization and trajectory modelling for source identification: a new look at Indian Ocean Experiment ship observations. *Atmospheric Environment* 42(20), 4836–4852

Bond, T. C., D. G. Streets, K. F. Yarber, S. M. Nelson, J.-H. Woo, and Z. Klimont (2004), A technology-based global inventory of black and organic carbon emissions from combustion. *Journal of Geophysical Research* 109, D14203, doi:10.1029/2003JD003697.

Borderieux, S., Wu, C-Y., Bonzongo, J-C., Powers, K., 2004. Control of Elemental Mercury Vapor in Combustion Systems Using Fe₂O₃. *Nanoparticles, Aerosol and Air Quality Research* 4 (1), 74–90.

Brigden, K., Labunska, I., Stringer, R., 2000. Identification of organic pollutants and heavy metal contaminants in filter ash collected from the Siderca primary and secondary steel smelter, Campana, Argentina. Technical note 20/00. <http://www.greenpeace.to/publications/Siderca%20report.pdf>. Accessed 08/03/13.

Callen, M. S., de la Cruz, M. T., Lopez, J. M., Navarro, M. V., Mastral, A. M., 2009. Comparison of receptor models for source apportionment of the PM10 in Zaragoza, Spain. *Chemosphere* 76, 1120-1129.

Castanho, A. D. A., Artaxo, P., 2001. Wintertime and summertime Sao Paulo aerosol apportionment study. *Atmospheric Environment* 35, 4889–4902.

573 Chan, K. C, Yao, X., 2008. Air pollution in mega cities in China. *Atmospheric Environment* 42, 1-
574 42.

575 Chavent, M., Guegan, H., Kuentz, V., Patouille, B., Saracco, J., 2007. Apportionment of Air
576 Pollution by Source at a French Urban Site. *CS-BIGS* 1(2), 119-129.
577

578 Cetin, B., Yarkin, S., Bayram, A., Odabasi, M., 2007. Ambient concentrations and source
579 apportionment of PCBs and trace elements around an industrialized area in Izmir, Turkey.
580 *Chemosphere* 69, 1267-1277.
581

582 Charron, A., Harrison, R. M., 2005. Fine ($PM_{2.5}$) and coarse ($PM_{2.5-10}$) particulate matter on a
583 heavily trafficked London highway: sources and processes. *Environmental Science and Technology*
584 39, 7768–7776.
585

586 Chio, C-P., Cheng, M-T., Wang, C-F., 2004. Source apportionment to PM_{10} in different air quality
587 conditions for Taichung urban and coastal areas, Taiwan. *Atmospheric Environment* 38, 6893-6905.
588

589 Chung, Y-S, Kim, S-H., Moon, J-H., Kim, Y-J, Lim, J-M, Lee, J-H., 2006. Source identification
590 and long-term monitoring of airborne particulate matter ($PM_{2.5}$ / PM_{10}) in an urban region of
591 Korea. *Journal of Radioanalytical and Nuclear Chemistry* 267 (1), 35-48.
592

593 Connell, D. P., Winter, S. E., Conrad, V. B., Kim, M., Crist, K. C., 2006. The Steubenville
594 Comprehensive Air Monitoring Program (SCAMP): Concentrations and solubilities of $PM_{2.5}$ trace
595 elements and their implications for source apportionment and health research. *Journal of the Air &*
596 *Waste Management Association* 56:1750–1766.
597

598 Dall'Osto, M., Booth, M. J., Smith, W., Fisher, R., Harrison, R. M. (2008a). Study of the size
599 distributions and the chemical characterization of airborne particles in the vicinity of a large
600 integrated steelworks. *Aerosol Science Technology* 42: 981–991.

601 Ebert, M., Müller-Ebert, D., Benker, N., Weinbruch, S., 2012. Source apportionment of aerosol
602 particles near a steel plant by electron microscopy. *Journal of Environmental Monitoring* 14, 3257-
603 3266.
604

605 EEA, 2012. European Union emission inventory report 1990–2010 under the UNECE Convention
606 on Long-range Transboundary Air Pollution (LRTAP). EEA Technical report No 8/2012
607

608 Fauser, P., Saarinen, K., Aasestrand, K., Danielsson, H., 2011. Emissions of Mercury, PAHs,
609 Dioxins and PCBs Related to NFR 3: Solvent and Other products use in Nordic countries. Nordic
610 Councils of Ministers, Copenhagen. TemaNord 2011: 549p. ISBN-978-92-893-2246-1.
611

612 Farao, C., Canepari, S., Perrino, C, Harrison, R.M., 2014. Sources of PM in an Industrial Area:
613 Comparison between Receptor Model Results and Semiempirical Calculations of Source
614 Contributions . *Aerosol and Air Quality Research*, In Press.
615

616 Garcia, J. H., Li, W-W., Cárdenas, N., Arimoto, R., Walton, J., Trujillo, D., 2006. Determination of
617 $PM_{2.5}$ sources using time-resolved integrated source and receptor. *Chemosphere* 65, 2018–2027.
618

619 Gildemeister, A. E., Hopke, P. K., Kim, E., 2007. Sources of fine urban particulate matter in
620 Detroit, MI. *Chemosphere* 69, 1064–1074.
621

622 Gupta, A. K., Karar, K. Srivastava, A., 2007. Chemical mass balance source apportionment of
623 PM10 and TSP in residential and industrial sites of an urban region of Kolkata, India. *Journal of*
624 *Hazardous Materials* 142, 279–287.

625

626 Hammond, D. M., Dvonch, J. T., Keeler, G. J., Parker, E. A., Kamal, A. S., Barres, J. A., Fuyen, Y.
627 P., Brake-Caldwell, W., 2008. Sources of ambient fine particulate matter at two community sites in
628 Detroit, Michigan. *Atmospheric Environment* 42:720–32.

629

630 Harrison, R. M., Beddows, D. C. S., Dall'Osto, M., 2011. PMF analysis of wide-range particle size
631 spectra collected on a major highway, *Environmental Science Technology* 45, 5522–5528.

632

633 Harrison, R. M., Beddows, D. C. S., Hu, L., Yin, J., 2012. Comparison of methods for evaluation of
634 wood smoke and estimation of UK ambient concentrations. *Atmospheric Chemistry and Physics* 12
635 (17), 8271–8283.

636 Harrison, R. M., Jones, A. M., Lawrence, R. G., 2003. A pragmatic mass closure model for airborne
637 particulate matter at urban background and roadside sites. *Atmospheric Environment* 37 (35), 4927–
638 4933.

639 Harrison, R. M., Yin, J., 2000. Particulate matter in the atmosphere: which particle properties are
640 important for its effects on health? *Science of the Total Environment* 249 (1-3), 85–10.

641

642 Hays, M. D., Fine, P. M., Geron, C. D., Kleeman, M. J., Gullett, B. K., 2005. Open burning of
643 agricultural biomass: physical and chemical properties of particle-phase emissions. *Atmospheric*
644 *Environment* 39 (36), 6747–6764.

645

646 Heal, M. R., Hibbs, L. R., Agius, R. M., Beverland, I. J., 2005. Total and water-soluble trace metal
647 content of urban background PM10, PM2.5 and black smoke in Edinburgh, UK. *Atmospheric*
648 *Environment* 39, 1417–1430.

649

650 Henry, R. C., 1997. History and Fundamentals of Multivariate Air Quality Receptor Models,
651 *Chemometrics and intelligent laboratory systems* 37, 525–530.

652 Henry R. C., 2002. Multivariate receptor models- current practices and future trends. *Chemometrics*
653 *and intelligent laboratory systems* 60, 43– 48.

654

655 Heo, J-B., Hopke, P.K., Yi, S.M., 2009. Source apportionment of PM2.5 in Seoul, Korea *Atmos.*
656 *Chem. Phys.*, 9, 4957–4971.

657

658 Hien, P. D., Binh, N. T., Truong, Y., Ngo, N. T., Sieu, L. N., 2001. Comparative receptor
659 modelling study of TSP, PM2 and PM2-10 in Ho Chi Minh City. *Atmospheric Environment* 35,
660 669–2678.

661

662 Hleis, D., Fernandez-Olmo, I., Ledoux, F., Kfoury, K, Courcot, L., Desmonts, T., Courcot, D.,
663 2013. Chemical profile identification of fugitive and confined particle emissions from an integrated
664 iron and steelmaking plant. *Journal of Hazardous Materials* 250–251, 246– 255.

665 Hopke, P. K., 2003. Recent development in receptor modelling. *Journal of Chemometrics* 17, 255–
666 265.

667

668 Jang, E., Alam, M.S., Harrison, R.M., 2013, Source apportionment of polycyclic aromatic
669 hydrocarbons in urban air using positive matrix factorization and spatial distribution analysis.
670 *Atmos. Environ* 79, 271–285

671 Johansson, M. T., Söderström, M., 2011. Options for the Swedish steel industry e Energy
672 efficiency measures and fuel conversion. *Energy* 36, 191-198.

673 Karar, K., Gupta, A. K., 2007. Source apportionment of PM₁₀ at residential and industrial sites of
674 an urban region of Kolkata, India. *Atmospheric Research* 84, 30–41.

675

676 Karnae, S., John, K., 2011. Source apportionment of fine particulate matter measured in an
677 industrialized coastal urban area of South Texas. *Atmospheric Environment* 45, 3769-3776.

678

679 Kim, M., Deshpande, S. R., Crist, K. C., 2007. Source apportionment of fine particulate matter
680 (PM_{2.5}) at a rural Ohio River Valley site. *Atmospheric Environment* 41, 9231-9243.

681

682 Kim, M-K, Jo, W-K., 2006. Elemental composition and source characterization of airborne PM₁₀ at
683 residences with relative proximities to metal-industrial complex. *International Archives of*
684 *Occupational and Environmental Health* 80, 40-50.

685

686 Kothai, P., Saradhi, I.V., Pandit, G.G., Markwitz, A., Puranik, V.D., 2011. Chemical
687 characterization and source identification of particulate matter at an urban site of Navi Mumbai,
688 India. *Aerosol and Air Quality Research*, 11, 560-569.

689

690 Kumar, A. V., Patil, R. S., Nambi, K. S. V., 2001. Source apportionment of suspended
691 particulate matter at two traffic junctions in Mumbai, India. *Atmospheric Environment* 35, 4245–
692 4251.

693

694 Lim, J-M., Lee, J-H., Moon, J-H., Chung, Y-S., Kim, K-H., 2010. Source apportionment of PM₁₀ at
695 a small industrial area using positive matrix factorization. *Atmospheric Research* 95, 88-100.

696

697 Lodhi, A., Ghauri, B., Khan, M.R., Rahman, S., Shafique, S., 2009. Particulate matter (PM_{2.5})
698 concentration and source apportionment in Lahore. *Journal of the Brazilian Chemical Society*, 20,
699 1811-1820.

700

701 Machemer, S.D., 2004. Characterization of airborne and bulk particulate from iron and steel
702 facilities. *Environmental Science and Technology* 38, 381-389.

703

704 Manahan, T. E., 2009. *Environmental Chemistry*, (ninth ed.), CRC Press, New York.

705

706 Mansha, M., Ghauri, B., Rahman, S., Amman, A., 2012. Characterization and source apportionment
707 of ambient air particulate matter (PM_{2.5}) in Karachi. *Science of the total Environment* 425: 176-83.

708

709 Marcazzan, G. M., Ceriani, M., Valli, G., Vecchi, R., 2003. Source apportionment of PM₁₀ and
710 PM_{2.5} in Milan (Italy) using receptor modelling. *Science of the Total Environment* 317, 137–147.

711

712 Mazzei, F., D'Alessandro, A., Lucarelli, Nava, S., Prati, P., Valli, G., Vecchi, R., 2008.
713 Characterization of particulate matter sources in an urban environment. *Science of the total*
714 *Environment* 401, 81-89.

715

716 Moreno, T., Jones, T. P., Richards, R. J., 2004. Characterisation of aerosol particulate matter from
717 urban and industrial environments: examples from Cardiff and Port Talbot, South Wales, UK.
718 *Science of the Total Environment*, 334-335, 337-346.

719

720 Mukherjee, A. B., Zevenhoven, R., Bhattacharya, P., Sajwan, K. S., Kikuchi, R., 2008.
721 Mercury flow via coal and coal utilization by-products: A global perspective, *Resource*
722 *Conservation Recycling* 52, 571–591.

723 Ogulei, D., Hopke, P.K., Chalupa, D.C., Utell, M. J., 2007. Modelling source contributions to
724 submicron particle number concentrations measured in Rochester, NY. *Aerosol Science and*
725 *Technology* 41, 179–201.

726

727 Ogulei, D., Hopke, P. K., Zhou, L., Pancras, J.P., Nair, N., Ondov, J.M., 2006. Source
728 apportionment of Baltimore aerosol from combined size distribution and chemical composition
729 data. *Atmospheric Environment* 40, S396–S410.

730

731 Oravisjarvi, K., Timonen, K. L., Wiikinkoski, T., Ruuskanen, A. R. Heinanen, K., Ruuskanene, J.,
732 2003. Source contributions to PM_{2.5} particles in the urban air of a town situated close to a steel
733 works. *Atmospheric Environment* 37, 1013-1022.

734

735 Paatero, P., Tapper, U., 1994. Positive matrix factorization; A non-negative factor model with
736 optimal utilization of error estimates of data values. *Environmetrics* 5, 111-126.

737

738 Pacyna, E. G., Pacyna, J. M., 2002. Global emission of mercury from anthropogenic sources
739 in 1995. *Atmospheric Environment* 37, 149–165.

740

741 Pacyna, E. G., Pacyna, J. M., Steenhuisen, F., Wilson S., 2006. Global anthropogenic mercury
742 emission inventory for 2000. *Atmospheric Environment* 40, 4048–4063.

743

744 European Commission, 2001. Ambient air pollution by polycyclic aromatic hydrocarbons (PAH).
745 Office for Official Publications of the European Communities, Luxembourg

746

747 Pancras, J. P., Landis, M. S., Norris, G. A., Vedantham, R., Dvonch, J. T., 2013. Source
748 apportionment of ambient fine particulate matter in Dearborn, Michigan, using hourly resolved PM
749 chemical composition data. *Science of the Total Environment* 448, (15), 2–13.

750

751 Pant, P., Harrison, R. M., 2012. Critical review of receptor modelling for particulate matter: A case
752 study of India. *Atmospheric Environment* 49, 1-12.

753

754 Pant, P., Harrison, R. M., 2013. Estimation of the Contribution of Road Traffic Emissions to
755 Particulate Matter Concentrations from Field Measurements: A Review. *Atmospheric Environment*
756 77, 78-97.

757

758 Pirrone, N., Costa, P., Pacyna, J. M., Ferrara, R., 2001. Mercury emissions to the atmosphere
759 from natural and anthropogenic sources in the Mediterranean region. *Atmospheric Environment* 35,
760 2997-3006. doi: 10.1016/S1352-2310(01)00103-0.

761

762 Pope, C. A., Dockery, D. W., 2006. Health Effects of Fine Particulate Air Pollution: Lines that
763 Connect. *Journal of Air & Waste Management Association* 56, 709–742.

764

765 Poschl, U., 2005. *Atmospheric Aerosols: Composition, Transformation, Climate and Health Effects*.
766 *Angewandte Chemie International Edition* 44 (46), 7520-7540.

767

768 Prati, P., Zucchiatti, A., Lucarelli, F., Mando, P. A., 2000. Source apportionment near a steel
769 plant in Genoa (Italy) by continuous aerosol sampling and PIXE analysis. *Atmospheric*
770 *Environment* 34, 3149-3157.

771

772 Qin, Y., Oduyemi, K., 2003. Atmospheric aerosol source identification and estimates of source
773 contributions to air pollution in Dundee, UK. *Atmospheric Environment* 37, 1799-1809.

774

775 Querol, X., Alastuey, A., de la Rosa, J., Sánchez-de-la-Campa, A., Plana, F., Ruiz, C. R., 2002.
 776 Source apportionment analysis of atmospheric particulates in an industrialised urban site in south-
 777 western Spain. *Atmospheric Environment* 36 (19), 3113-3125.
 778
 779 Querol, X., Viana, M., Alastuey, A., Amato, F., Moreno, T., Castillo, S., Pey, J., de la Rosa, J.,
 780 Sánchez de la Campa, A., Artíñano, B., Salvador, P., García Dos Santos, S., Fernández-Patier, R.,
 781 Moreno-Grau, S., Negral, L., Minguillón, M.C., Monfort, E., Gil, J.I., Inza, A. Ortega, L.A.,
 782 Santamaría, J.M., Zabalza, J., 2007. Source origin of trace elements in PM from regional
 783 background, urban and industrial sites of Spain. *Atmospheric Environment* 41 (34), 7219-7231.
 784
 785 Querol, X., Zhuang, X., Alastuey, A., Viana, M., Lv, W., Wang, Y., Lopez, A., Zhu, Z., Wei, H.,
 786 Xu, S., 2006. Speciation and sources of atmospheric aerosols in a highly industrialized merging
 787 mega-city in Central China. *Journal of Environmental Monitoring* 8, 1049-1059.
 788
 789 Ravindra, K., Sokhi, R., Van Grieken, R., 2008. Atmospheric polycyclic aromatic hydrocarbons:
 790 source attribution, emission factors and regulation. *Atmos Environ*, 42, 2895-2921
 791
 792 Rehwagen, M., Muller, A., Massolo, L. Herbath, O. Ronco, A., 2005. Polycyclic aromatic
 793 hydrocarbons associated with particles in ambient air from urban and industrial areas. *Science of*
 794 *the Total Environment* 348 (1-3), 199-210.
 795
 796 Samara, C., Kouimtzis, Th., Tsitouridou, R., Kanias, G., Simeonov, V., 2003. Chemical mass
 797 balance source apportionment of PM₁₀ in an industrialized urban area of Northern Greece.
 798 *Atmospheric Environment* 37, 41-54.
 799
 800 Srivastava, A., Jain, V.K., 2008. Source apportionment of total suspended particulate matter in
 801 coarse and fine size ranges over Delhi. *Aerosol and Air Quality Research*, 8, 188-200.
 802
 803 Sternbeck, J., Sjödin, A., Andreasson, K., 2002. Metal emissions from road traffic and the influence
 804 of resuspension—results from two tunnel studies. *Atmospheric Environment* 36, 4735 –4744.
 805
 806 Streets, D. G., Gupta, S., Waldhoff, S. T., Wang, M. Q., Bond, T. C., Bo, Y., 2001. Black carbon
 807 emissions in China *Atmospheric Environment* 35, 4281–4296.
 808
 809 Taiwo, A.M., Beddows, D.C.S., Calzolari, G., Harrison, R.M., Lucarelli, F., Nava, S., Shi, Z., Valli,
 810 G., Vecchi, R., 2014. Receptor modelling of airborne particulate matter in the vicinity of a major
 811 steelworks site. *Science of the Total Environment*, 490, 488-500.
 812
 813 Themelis, N. J., Gregory, A. F., 2002. Mercury Emissions from High Temperature Sources in
 814 the NY/NJ Hudson- Raritan Basin, Proceedings of NAWTEC 10. American Society of Mechanical
 815 Engineers, 205–215.
 816
 817 Thorpe, A. and Harrison, R. M., 2008. Sources and properties of non-exhaust particulate matter
 818 from road traffic: a review. *Science of the Total Environment* 400, 270-282.
 819
 820 Thurston, G.D., Spengler, J. D. A., 1985. Quantitative assessment of source contributions to
 821 inhalable particulate matter pollution in metropolitan Boston. *Atmospheric Environment* 19, 9-25.
 822
 823 Tsai, J-H., Lin, K-H., Chen, C-Y., Ding, J-Y., Choa, C-G., Chiang, H-L., 2007. Chemical
 824 constituents in particulate emissions from integrated iron and steel facility. *Journal of Hazardous*
 825 *Materials* 147, 111-119.
 826

827 Tsai, J-H., Lin, K.H., Chen, C-Y., Lai, N., Ma, S.-Y., Chiang, H-L., 2008. Volatile organic
 828 compound constituents from an integrated iron and steel facility. *Journal of Hazardous Materials*
 829 157, 569–578.

830

831 Viana, M., Kuhlbusch, T.A.J., Querol, X., Alastuey, A., Harrison, R. M., Hopke, P. K., Winiwarter,
 832 W., Vallius, M., Szidat, S., Prévôt, A. S. H., Hueglin, C., Bloemen, H., Wåhlin, P., Vecchi, R.,
 833 Miranda, A. I., Kasper-Giebl, A., Maenhaut, W., Hitzenberger, R., 2008a. Source Apportionment of
 834 Particulate Matter in Europe: A Review of Methods and Results. *Aerosol Science* 39, 827-849

835

836 Viana, M., Pandolfi, M, Minguillo, M. C., Querol, X., Alastuey, A., Monfort, E., Celades, I., 2008b.
 837 Inter-comparison of receptor models for PM source apportionment: Case study in an industrial area.
 838 *Atmospheric Environment* 42, 320-3832.

839

840 Watson, J. G., Judith, T. Z., Chow, C., Eric, J. E., Fujita, M., Wilson, W. E., 2002. Receptor
 841 modeling application framework for particle source apportionment. *Chemosphere* 49, 1093–1136.

842

843 Watson, J. G., Chow, J. C., Houck, J. E., 2001. PM_{2.5} chemical source profiles for vehicle exhaust,
 844 vegetative burning, geological material, and coal burning in northwestern Colorado during 1995.
 845 *Chemosphere* 43 (8), 1141-1151.

846

847 Weitkamp, E. A., Lipsky, E. M., Pancras, P. J., Ondov, J. M., Polidori, A., Turpin, B. J., Robinson,
 848 A. L., 2005. Fine particle emission profile for a large coke production facility based on highly time-
 849 resolved fence line measurements. *Atmospheric Environment* 39, 6719-6733.

850

851 WHO, 2006. WHO Air quality guidelines: Global update 2005. World Health Organization,
 852 Copenhagen, Denmark.

853

854 Xue, Y-H., Wu, J-H., Feng, Y-C., Dai, L., Bi, X-H, Li, X., Zhu, T., Tang, S-B., Chen, M-F., 2010.
 855 Source characterization and apportionment of PM₁₀ in Panzhihua, China. *Aerosol and Air Quality*
 856 *Research* 10, 367-377.

857

858 Yatkin, S., Bayram, A., 2008. Source apportionment of PM₁₀ and PM_{2.5} using positive matrix
 859 factorization and chemical mass balance in Izmir, Turkey. *Science of the Total Environment*, 390,
 860 109-123.

861

862 Yin, J. Harrison, R. M., Chen, Q., Rutter, A., Scauer, J. J., 2010. Source apportionment of fine
 863 particles at urban background and rural sites in the UK atmosphere. *Atmospheric Environment* 44,
 864 841-851.

865

866 Zhang, R., Jing, J., Tao, J., Hsu, S.C., Wang, G., Cao, J., Lee, C.S.L., Zhu, L., Chen, Z., Zhao, Y.,
 867 Shen, Z., 2013. Chemical characterization and source apportionment of PM_{2.5} in Beijing: seasonal
 868 perspective. *Atmos Chem Phys*, 13, 7053-7074.

869

870 Zhang, X., Hecobian, A., Zheng, M., Frank, N. H., Weber, R. J., 2010. Biomass burning impact on
 871 PM 2.5 over the southeastern US during 2007: integrating chemically speciated FRM filter
 872 measurements, MODIS fire counts and PMF analysis. *Atmospheric Chemistry and Physics* 10(14),
 873 6839-6853.

874

875 Zeng, F., Shi, G-L., Li, X., Feng, Y-C., Bi, X-H., Wu, J.H., Xue, Y-H., 2010. Application of a
 876 combined model to study the source apportionment of PM₁₀ in Taiyuan, China. *Aerosol and Air*
 877 *Quality Research* 10, 177–184.

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TABLE LEGENDS

- Table 1:** Contribution of each trace metal from industrial processes to total emissions and their major sources in the UK based on 2009 United Kingdom National Atmospheric Emissions Inventory.
- Table 2:** Results of Source Apportionment (SA) with different receptor models at industrial sites.
- Table 3:** Source Apportionment with different receptor models at residential sites (selected studies) with a reported industrial contribution.

FIGURE LEGENDS

- Figure 1:** Source profiles of charcoal manufacturing, sinter plant and BOF plant (with electrostatic precipitator). From US EPA SPECIATE database.
- Figure 2:** Scatter plots of PMF factor profiles (in percentage) from published studies versus USEPA SPECIATE source profiles for (a) general steel production, (b) sinter plant, (c) coke plant and (d) blast furnace. In Fig. 2a, Gildemeister et al. (2007) (1): site 1-Allen Park site; Gildemeister et al. (2007) (2): Dearborn site at Detroit industrial area, USA; PMF profile in Taiwo et al. (2014) (Fig. 2a) was a mixed factor comprising Steel 2 (BOS) and Steel 4 (sinter plant); PMF factor profiles in Gildemeister et al. (2007) were kindly provided by the authors; PMF profiles of iron/steel factor from Pancras et al. (2013) were estimated (so carries small subjective error) from concentration of each element (in ng m^{-3}) in Fig. 3 in the original paper and the apportioned iron/steel factor concentration of $0.36 \mu\text{g m}^{-3}$ (Table 4 of the original paper). PMF factor profiles of sinter plant (Fig. 2b) and coke dust (Fig. 2c) are from Alleman et al., (2010) and that of blast furnace is from Taiwo et al. (2014). The USEPA SPECIATE blast furnace profile: PM (0-38 μm) from kish graphite from blast furnace process in iron and steel manufacturing. Ni and Ba made negligible contribution to factor Steel 2 and 4 in Taiwo et al. (2014) so were not included in comparison (Fig. 2a); similarly Cr made negligible contribution to factor Steel 1 in Taiwo et al. (2014) so was not included in comparison (Fig. 2d). Please note that some elements were reported in USEPA SPECIATE source profiles but not in PMF factor profiles; and vice versa. In those cases, the data could not be shown in the figures.
- Figure 3:** Regression plots of USEPA SPECIATE vs Tsai et al. (2007) source profiles for the sintering process in the steel industry.

923 **Table 1:** Contribution of each trace metal from industrial processes to total emissions and their
924 major sources in the UK based on 2009 United Kingdom National Atmospheric Emissions
925 Inventory.
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	Total Emissions (tonnes)	Industrial contribution %	Major sources
As	13	93	Treated wood for industrial combustion; metal production; public electricity and heat production
Cd	2	78	Non-ferrous metal production and iron and steel manufacture (as well as other forms of industrial combustion), energy production (include a significant proportion from waste combustion and fuel oil combustion for electricity generation)
Cr	26	89	Coal combustion, iron and steel production in integrated works and in electric arc furnaces and the production of chromium based chemicals
Cu	52	49	Metal production, combustion of lubricants in industry and coal combustion
Pb	60	87	Metal production and combustion of lubricants in industry
Hg	7	99	Iron and steel production processes, public electricity and heat production, waste incineration, the manufacture of chlorine in mercury cells, coal and other forms of industrial combustion
Ni	83	54	Combustion of heavy fuel oil
Se	31	92	Glass production and combustion for public electricity and heat production
V	477	21	Fuel oil combustion
Zn	339	72	Metal production and combustion in industry

927 Note: this table is adapted from UK emissions of air pollutants 1970 to 2009 by AEA (2011):
928 available at http://uk-air.defra.gov.uk/reports/cat07/1401131501_NAEI_Annual_Report_2009.pdf
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Table 2: Results of Source Apportionment (SA) with different receptor models at industrial sites.

Ref.	Study Area	Setting of Study Area and population	Method of SA	Parameter used for SA	Types of Industry	PM Size	Marker elements for industries	PM conc.: $\mu\text{g}/\text{m}^3$	Source Contribution (%)							
									Combustion (coal, wood, oil, gas, biomass),*	Marine or other salt source (e.g. road salt)	Vehicle emission (exhaust & non-exhaust)	Secondary aerosol, Regional or long range transport	Crustal matter **	Industry	Incineration or waste Incineration	Un-explained or Others
Hien et al., 2001	Ho Chi Minh City, Vietnam	Industrial, commercial 4.5 million	PCFA	Metals	Small factories	TSP PM _{2.5} PM ₁₀	Ce, Co, As, Cr, Pb, Sb	74 32 16	11 16 18	18	6 17 17	25	77 33 27	4 2 13		2 14
Kumar et al., 2001	Mumbai India	Industrial/ Residential	PCA	Metals, OC/EC, ions, NO ₂	Medium scale industries	SPM, likely to be TSP	Cu, Mn, Ni	1032-1176	6-11	15	15-18		33-41	6-8		15-17
Oravisiarvi et al., 2003	Raahe, Finland	Industrial, population-17,000	PCA	Metals, Ions	Steel/Mechanical engineering Works	PM _{2.5}	Mn, F, Zn, Fe, Ca, Cd, K, Na, Pb, Cl Cu, Mo, Ni, Cr	10				44	7	14		35
Samara et al., 2003	Thessaloniki, Greece	Industrial	CMB	Metals, ions, PAHs	Steel, Oil refineries, metallurgy, cement	PM ₁₀	V, Fe, Zn, Pb, Cl, Ca, Na, SO ₄ , NO ₃	-	8-28		47-64		19-29	0-7		-

Chung et al., 2006	Urban Daehwa, Korea	Industrial 1.4 million	PMF	Metals	Soap, cosmetics, metallurgy, plastic, chemicals	PM _{2.5} PM _{2.5-10}	Al, Cl, Cr, Ti, Fe, Co, As, Cu	10 23	36 24	3 9	24 5		2 55	28 2	9 5	
Connell et al., 2006	Steubenville, USA	Industrial, 132,000	PMF	Metals, anions	Coke, Metal Smelting and processing	PM _{2.5}	Fe, Mn, Zn, Mg, As, Zn, Cu, Cd, Pb	18	3		20	57	6	13		
Kim and Jo, 2006	Pohang, Korea	Industrial	PCA	Metals	Metal industries	PM ₁₀	Cu, Mn, Tl,	76			9		35	10	18	28
Ogulei et al., 2006	Baltimore Supersite, USA	Industrial	PMF	Metals, ions, OC/EC, Gases	Steel, Automotive painting	PM _{2.5}	Fe, Cu, Pb		44		10	33	3	2	8	
Querol et al., 2006	Changqian, Wuhan City, China	Industrial 9 million	PCA	Metals, ions, OC/EC	Steel, Petrochemical	PM ₁₀	Cd, Bi, Rb, As, Cu, Pb, Sb, Sn and K	197	20		10	16	34	15		5
Gupta et al., 2007	Kolkata, India	Industrial	CMB	Metals, PAHs, Ions	Metal, electroplating	PM ₁₀ TSP			34 17		47 7		1 52	1		17 24
Cetin et al., 2007	Aliaga, Turkey	Industrial	CMB	Metals, PAHS	Steel	PM ₁₀	Zn, Fe, Pb, Mn, Cd	87 w 60 s	25 w 7 s	1 w	7 w		12 s	70w 55 s		26 s
Karar and Gupta, 2007	Kolkata, India	Industrial	PCA-APCS	Metals, PAHs, TC, OC, Ions		PM ₁₀	Cr	197	29		45	1		18		7

Gildemeister et al., 2007	Dearborn, Detroit, USA	Industrial	PMF	Metals, Anions, OC/EC	Metallurgy, Steel, Oil refinery	PM _{2.5}	Fe, K, Mn, Zn, Ca	19	8	4	30	43	12	12		
Chavent et al., 2007	Anglet, South West, France	Industrial 170, 000	Factor Analysis	Metals	Steel	PM _{2.5}	Zn, Pb		78	5	9		6	3		
Mazzei et al., 2008	Cornigliano, Genoa, Italy	Industrial	PMF	Metals	Steel	PM ₁ PM _{2.5} PM ₁₀	Fe, Mn, Zn, Pb	18 19 42	58 40 -	- - 17	9 17 15	11 7 36	7 17 8	7 20 23		6 3 1
Viana et al., 2008b	Castello, Spain	Industrial	PCA PMF CMB	Metals, OC/EC, Ions	Ceramic, petrochemical, organic chemicals	PM ₁₀	As, Zn, Pb, Cs, Tl, Zr, Hf, Ce	34		3	10 10 13	25 18	12	48 32 47		42 33 7
Yatkin and Bayram, 2008	Izmir, Turkey	Industrial, 3 millions	PMF CMB	Metals	Iron & steel, cement, minerals	PM _{2.5}	Zn, Pb, Mn, Fe, V, Ni	64	7-11	2-3	15 80-81		1 1-3	1-1		70
Amato et al., 2009	Bacelonia, Spain	Industrial, 1.6 million	ME-2	Metals, ions, total carbon	Asphalt, Ferrous and non ferrous smelters, Cement Power plants	PM ₁ PM _{2.5} PM ₁₀	Pb, Zn, Fe, Mn, Cd	17 28 40	8 6 5	0 3 10	36 32 21	48 26 24	3 21 42	2 2 1		
Amato et al., 2009	Bacelonia, Spain	Industrial, 1.6 million	PMF	Metals, ions, total carbon	Asphalt, Ferrous and non ferrous smelters, Cement Power plants	PM ₁ PM _{2.5} PM ₁₀	Pb, Zn, Fe, Mn, Cd	17 28 40	8 7 6	0 3 11	30 29 28	53 45 25	0 14 31	3 3 2		

Lodhi et al., 2009	Lahore, Pakistan	Industrial, Urban, 6.6 million	PMF	Metals, ions	Steelworks, Power plants	PM _{2.5}	Co, Cr, Fe, Mo, Ni, Sn				5	51	18	8		18
Alleman et al, 2010	Dunkirk, France	Industrial 210, 000	PMF	Metals	Mixed Industrial zone	PM ₁₀	Fe, Ca, Si, Mo, As, Cd, Pb, V, Ni, Ti, Zn			12	15		24	37		12
Lim et al., 2010	Daejeon, Korea	Industrial 1.45 million	PMF	Metals, ions	Metallurgy, plastic, chemicals, cement	PM ₁₀	As, In, Cl, Fe, K, SO ₄ , NH ₄ , Pb	86	6		9	23	48	7		7
Xue et al., 2010	Panzhihua, China	Urban/Industrial	NCMB	Metals, Anions	Iron & steel, metallurgy	PM ₁₀	TC, V, Ti, Cr, Mn,	131-179	26		14	23	11	20		8
Zeng et al., 2010	Taiyuan, China	Industrial	PCA/MLR	Metals, ions, OC	Steel, Construction, chemical, energy	PM ₁₀	Co, Cr, Fe, Mo, Ni, Sn	305	18		13	16	38	12		3
Mansha et al., 2012	Karachi, Pakistan	Industrial 14 million	PMF	Metals, ions	Steel, Cement, Textile, Refineries, Petrochemical	PM _{2.5}	Co, Cr, Fe, Mo, Ni, Sn	84			19	12	16	53		
Pancras et al., 2013	Dearborn, Michigan, USA	Industrial	PMF	Metals, ions, OC/EC	Metallurgy, Steel, Oil refinery	PM _{2.5}	Se, SO ₂ , Fe, Mn, Pb, Cu, Zn, K, Rb	16	4		8	54	13	7	2	
Pancras et al., 2013	Dearborn, Michigan, USA	Industrial	Ummix	Metals, ions, OC/EC	Metallurgy, Steel, Oil refinery	PM _{2.5}	SO ₂ , Fe, Cu, K, Rb	16	3		28	60		14	1	

Farao et al., 2014	Ferrara, Po Valley, Italy	Industrial 132, 000	PMF	Metals, ions, OC/EC		PM ₁₀ PM _{2.5}	As, Cd, Pb, Tl, Zn			5 2	13	40	20 20	10 4		56 27
Taiwo et al., 2014	Port Talbot, South Wales, UK	Industrial 35, 000	ME-2	Metals, ions	Steel	PM ₁₀ PM _{2.5-10} PM _{2.5}	Fe, Mn, Ca, Ni, Cd, Pb, Zn	8 12 20		20 30 28	13 16 16	20 13		14 31 23		33 10 33

Note: NCMB-Nested chemical mass balance, CPF- Conditional Probability Function, PSCF- Potential Source Contribution Function. LMR-Least Multiple Regression, W-winter, S-summer; Sometimes two or more sources were grouped as one factor for example, in Kim and Jo (2006), Lim et al. (2010), Lodhi et al. (2009), Yarkin and Bayram (2008) and Viana et al. (2008b), in which the factor will be counted as “others”. For these reasons, readers are strongly advised to read the original articles for more details

*, Different authors used different definition of this combustion source for example, coal, wood, oil, and gas combustion, and biomass burning; they were grouped together for simplicity in this review. **, this category includes all crustal element based source, for example, soil dust, road dust, cement or minerals.

Table 3: Source apportionment with different receptor models at residential sites (selected studies) with a reported industrial contribution.

Authors	Study Area	Setting of Study Area	SA Method	Parameter	PM Size	Marker element for industry	PM conc. ($\mu\text{g}/\text{m}^3$)	Sources Contribution (%)							
								Combustion, coal, wood, oil, (cooking)	Marine aerosol	Traffic (Exhaust and non-exhaust)	Secondary aerosol	Crustal matter	Industry	Waste, Incineration	Others
Castanho, and Artaxo, 2001	Sao Paulo, Brazil	Residential 5.5 millions	APFA	Metals, OC, EC	PM _{2.5}	Zn, Mn, Pb, Fe, Ni, S	30 w, 15 s	19 21		28 24	23 17	25 31	5 6		
Almeida et al., 2005	Bobadela, Portugal	Residential	PCA/MLRA	Metals, OC, BC, Anions	PM _{2.5} PM _{2.5-10}	Zn, Cu, Sb, Pb	24 16	8 5	8 47	22	25 15	16 20	0.2 0.4		21 13
Alastuey et al., 2007	Tarragona harbour, Spain	Harbour	PCA-MLRA	Metals, Anions	PM ₁₀	V, Ni, Mn, Co	40		13	34		17	12		24
Kim et al., 2007	Ohio River Valley, USA	Residential	PMF	Metals, OC, EC, Ions	PM _{2.5}	Fe, Mn, Ca, SO ₄ , Zn, Pb	14			2.5	69	2.5	6		20
Srivastava and Jain, 2008	Delhi, India	Residential 14 millions	CMB	Metals	>1.6 μm <1.6 μm	Fe, Mn, Cu, Cr, Ni, Pb				29 62		68 36	3 2		
Srivastava and Jain, 2008	Delhi, India	Residential 14 millions	PCA	Metals	>1.6 μm <1.6 μm					23 86		68 10			
Yatkin and Bayram, 2008	Izmir, Turkey	Sub-urban, 3 millions	PMF	Metals	PM _{2.5}	Zn, Pb, Mn, Fe, V, Ni	24		4	12		9			75
Yatkin and Bayram, 2008	Izmir, Turkey	Sub-urban, 3 millions	CMB	Metals	PM _{2.5}	Zn, Pb, Mn, Fe, V, Ni	20 w 29 s	13 10	6 10	71 63		8 15	1 2		0.4 0.1
Callen et al., 2009	Zaragora, Spain	Urban	PCA-APCS Unmix PMF	Metals, OC, EC, Ions, PAHs, NH ₄	PM ₁₀		32	13 12	4 10 7	15 7		56 65 40			27 9 34

Zhang et al., 2013	Beijing, China	urban	PMF	Metals, anions, OC/EC	PM _{2.5}	OC, EC, Zn, Mn, and Cr	135	30			26	15	25		4
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Note: W-winter, S-summer; Sometimes two or more sources were grouped as one factor, in which the factor will be counted as “others”. For these reasons, readers are strongly advised to read the original articles for more details; Different authors used different definition of this combustion source for example, coal, wood, oil, and gas combustion, and biomass burning; they were grouped together for simplicity in this review

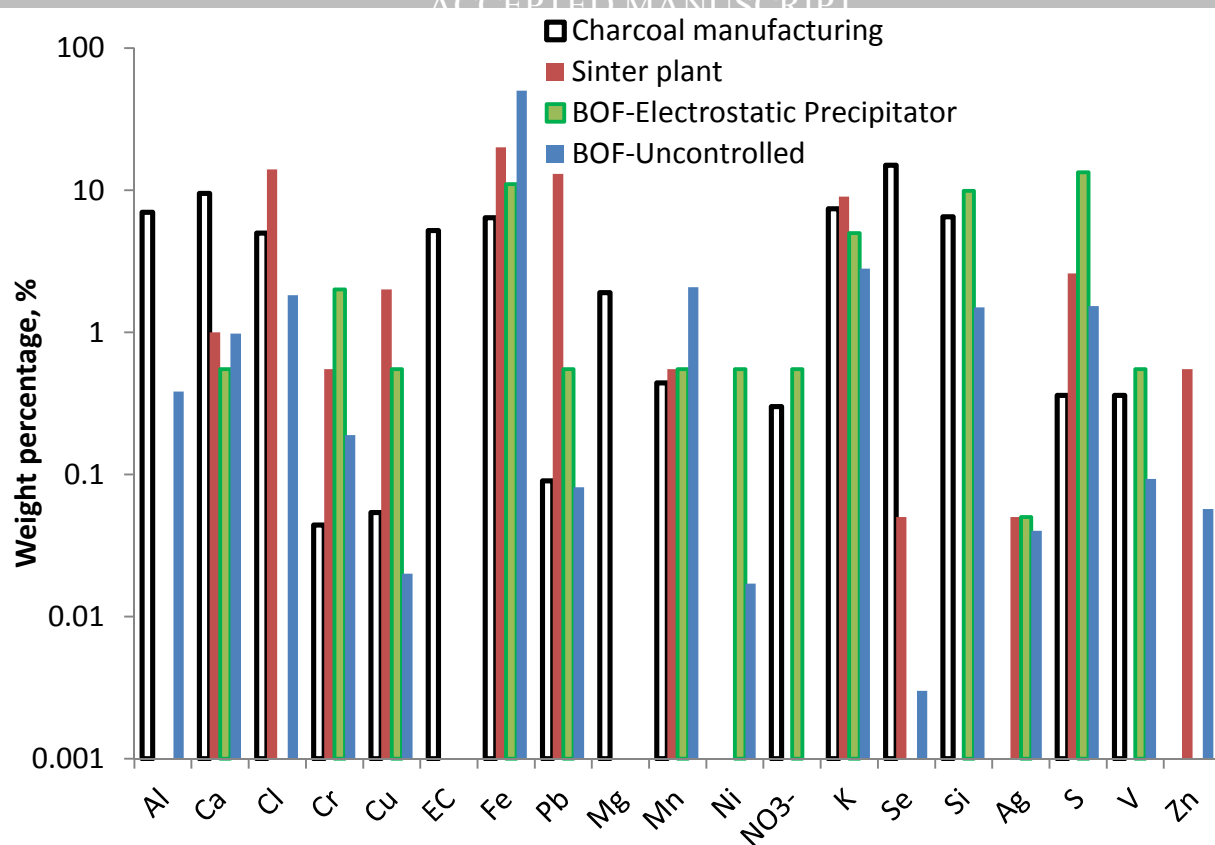
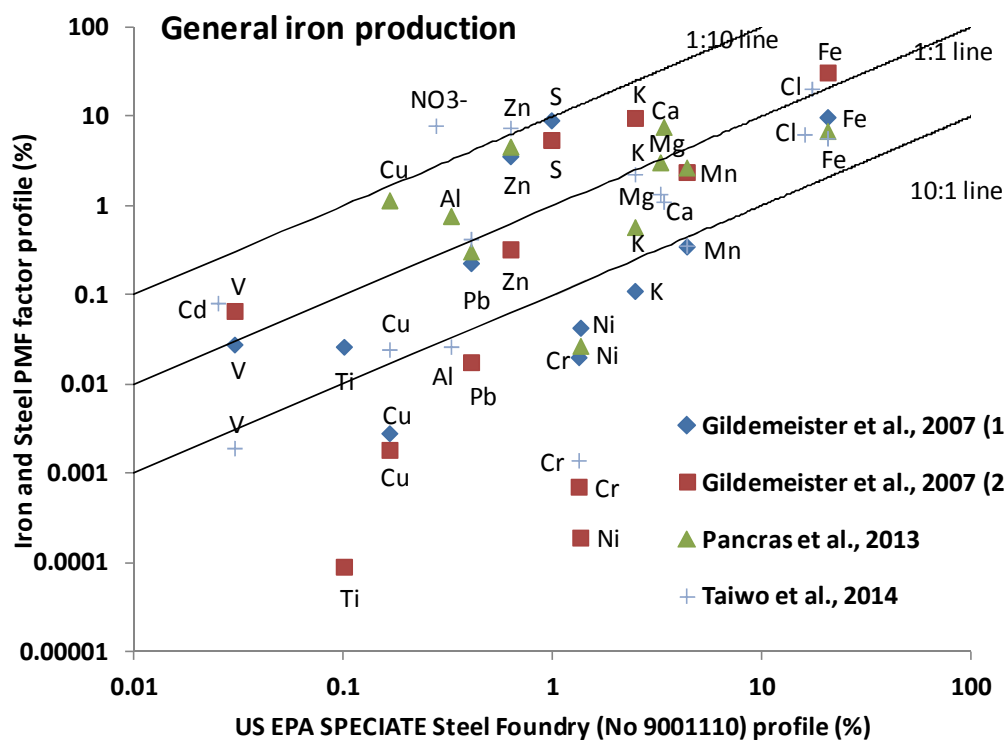
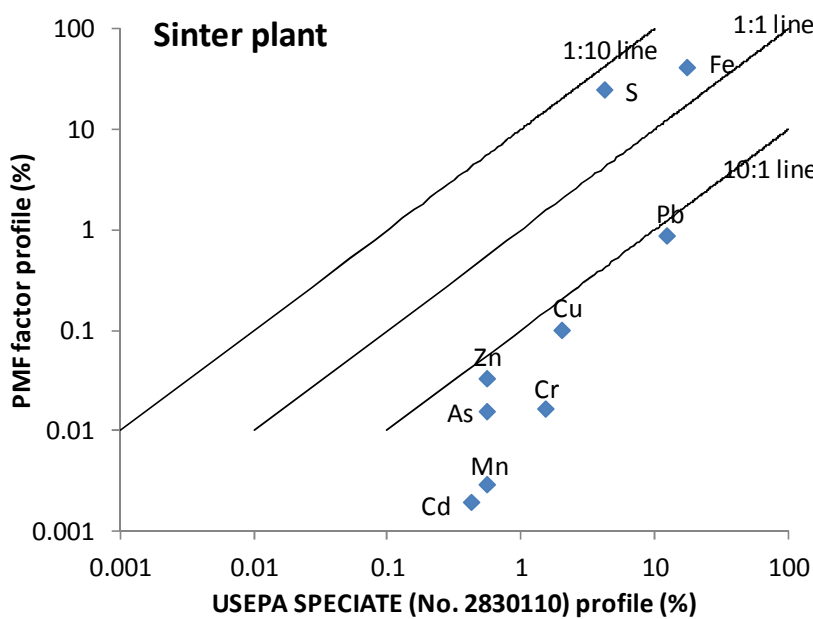


Figure 1: Source profiles of charcoal manufacturing, sinter plant and BOF plant (with electrostatic precipitator and uncontrolled). From US EPA SPECIATE database.



(a)



(b)

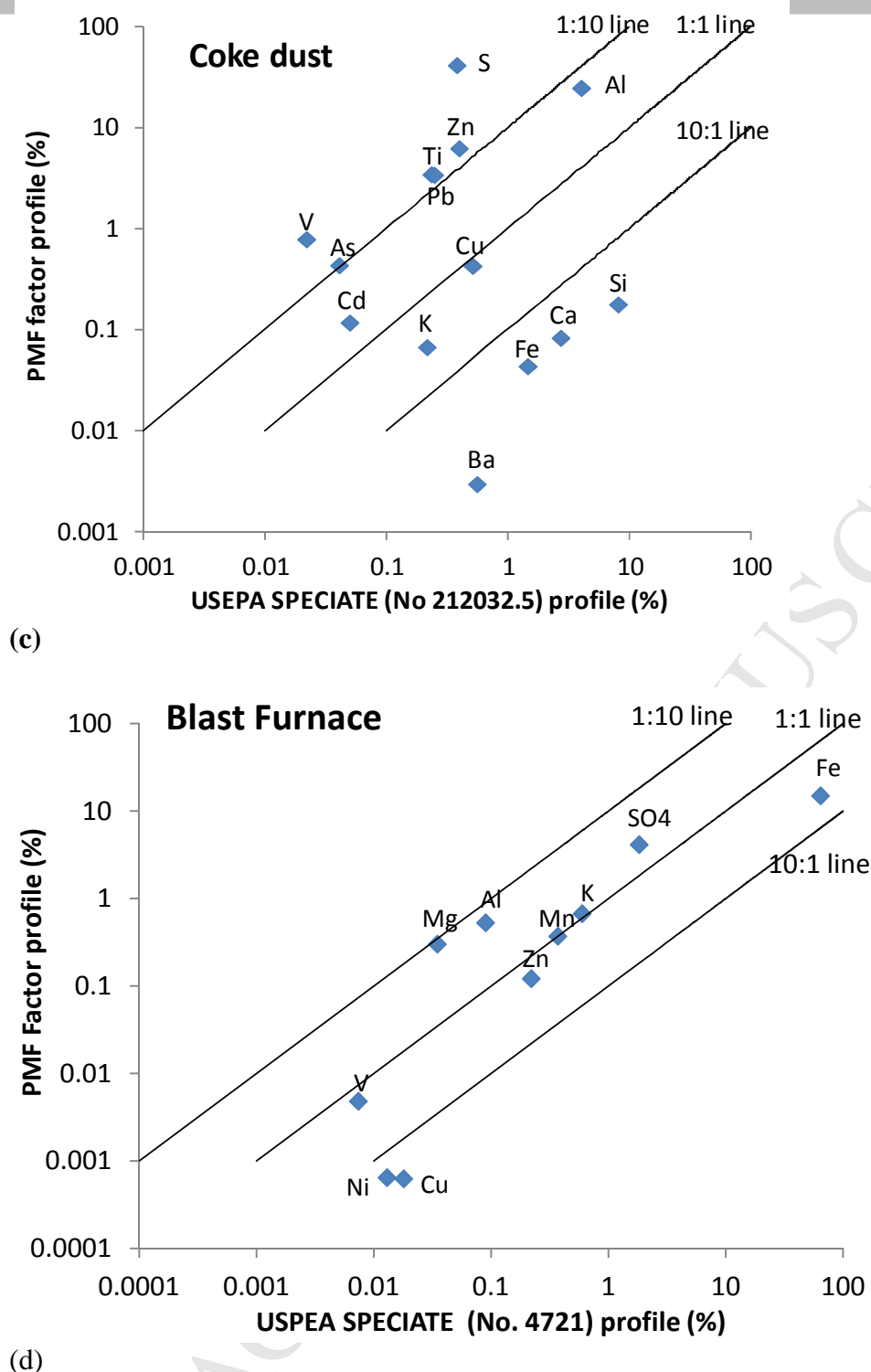


Figure 2: Scatter plots of PMF factor profiles (in percentage) from published studies versus USEPA SPECIATE source profiles for (a) general steel production, (b) sinter plant, (c) coke plant and (d) and blast furnace. In Fig. 2a, Gildemeister et al. (2007) (1): site 1-Allen Park site; Gildemeister et al. (2007) (2): Dearborn site at Detroit industrial area, USA; PMF profile in Taiwo et al. (2014) (Fig. 2a) was a mixed factor comprising Steel 2 (BOS) and Steel 4 (sinter plant); PMF factor profiles in Gildemeister et al. (2007) were kindly provided by the authors; PMF profiles of iron/steel factor from Pancras et al. (2013) were estimated (so carries small subjective error) from concentration of each element (in ng m^{-3}) in Fig. 3 in the original paper and the apportioned iron/steel factor concentration of $0.36 \mu\text{g m}^{-3}$ (Table 4 of the original paper). PMF factor profiles of sinter plant (Fig. 2b) and coke dust (Fig. 2c) are from Alleman et al., (2010) and that of blast

furnace (Fig. 2d) is from Taiwo et al. (2014). The USEPA SPECIATE blast furnace profile: PM (0-38 μm) from kish graphite from blast furnace process in iron and steel manufacturing. Ni and Ba made negligible contribution to factor Steel 2 and 4 in Taiwo et al. (2014) so were not included in comparison (Fig. 2a); similarly Cr made negligible contribution to factor Steel 1 in Taiwo et al. (2014) so was not included in comparison (Fig. 2d). Please note that some elements were reported in USEPA SPECIATE source profiles but not in PMF factor profiles; and vice versa. In those cases, the data could not be shown in the figures.

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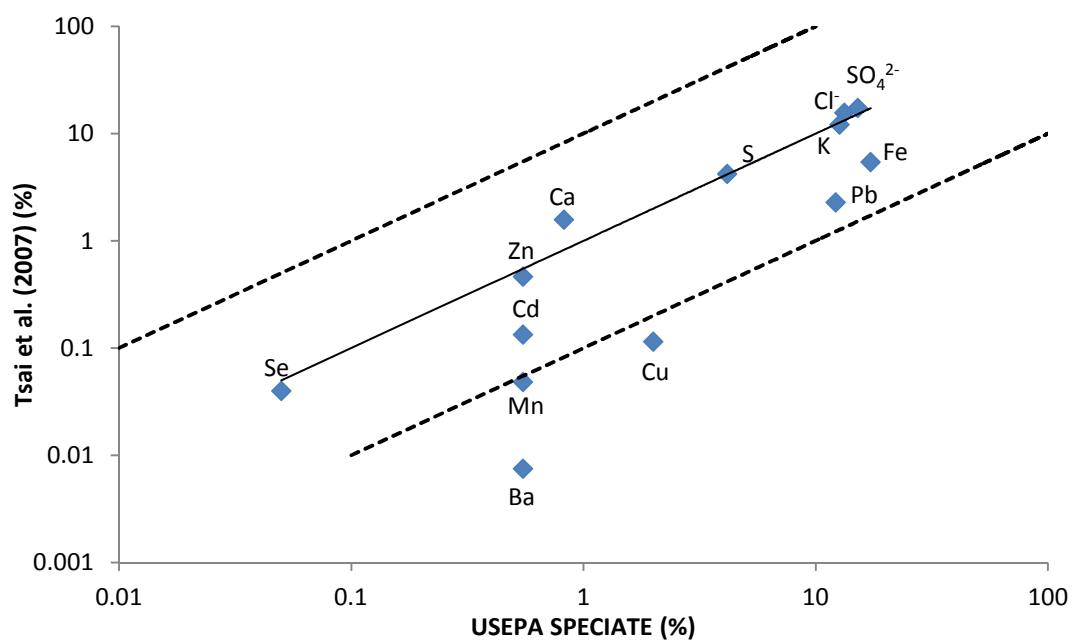


Figure 3: Regression plots of USEPA SPECIATE vs Tsai et al. (2007) source profiles for the sintering process in the steel industry.

A Review of Receptor Modelling of Industrially Emitted Particulate Matter

Adewale M. Taiwo, Roy M. Harrison and Zongbo Shi

HIGHLIGHTS

Industrial processes have been identified as an important source of airborne PM.

PM from different sites within the same industry may vary appreciably in composition.

PM from different processes within the same industrial site can differ substantially.

Local source profile measurements are needed for industrial PM source apportionment.